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EFFECTS OF ($\alpha$,n) CONTAMINANTS AND SAMPLE MULTIPLICATION ON STATISTICAL NEUTRON CORRELATION MEASUREMENTS

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## ABSTRACT

The complete formalism for the use of statistical neutron fluctuation measurements for the nondestructive assay of fissionable materials has been developed. This formalism includes the effect of detector deadtime, neutron multiplicity, random neutron pulse contributions from ($\alpha$,n) contaminants in the sample, and the sample multiplication of both fission-related and background neutrons.

## 1. Introduction

Detecting and analyzing the time correlations in the pulse trains from neutron detectors has become the established method for nondestructive assay of samples containing plutonium. Time correlations result from the multiplicity of neutrons emitted in a fission event or from chain correlated fission events in the case of multiplying samples. The presence of these time correlations provides for the possibility of separating the contributions to the pulse train of fission and random neutron emitting events. This separation has been attempted in various ways with varying degrees of success. The techniques have employed count time gates associated with variable deadtime circuits[1] or coincidence circuits[2] and contiguous time intervals associated with reduced variance logic circuits.[3,4] An experimental comparison[5] has been made of variable deadtime circuit and the shift register coincidence circuit,[6] and a theoretical comparison[7] has been made of the coincidence and reduced variance methods. In the former report, the shift register technique was shown to be a more accurate assay for plutonium than the variable deadtime technique over a wide range of sample sizes. In the latter report, the shift register technique was said to be difficult to analyze fully, and it is speculated that the reduced variance technique appears to extract more information from each gate period and might therefore offer more efficient use of the measurement time available, providing greater accuracy for the same overall time or less time consumed for the same accuracy. This speculation may result from the realization that the reduced variance method retains the history of the pulse population in an individual time gate whereas the coincidence methods average all of the individual time gate pulse populations.

We have shared this perception of the potential for obtaining more information from the pulse train using the reduced variance method, and were the first to accurately assay large multiplying plutonium metal samples (up to 4.1 kg) by making use of this method.[4] We continue to be motivated to explore the application of the reduced variance method to special nuclear material (SNM) assay and have expanded the formalism to include multiplying samples other than pure metals.

In the earlier paper,[4] the basic formalism for assay of plutonium using the reduced variance or neutron fluctuation technique was presented. This version of the formalism was limited to the special case in which all neutrons originate in fission events, either spontaneous or induced. It was shown that pure plutonium metal samples, even those for which multiplication was significant (>2), could be assayed using this technique. In addition, it was shown that the assay of nonmultiplying samples containing ($\alpha$,n) contaminants was possible.

Presented in this paper is the complete formalism for the use of neutron fluctuation measurements for plutonium assay. It accounts for sample multiplication and ($\alpha$,n) contaminants in addition to spontaneous fissions in the sample. The effect of detector deadtime on the measurement is carried completely through this formalism. Measurements using nonmultiplying $^{252}Cf$ and ($\alpha$,n) sources are discussed and the results used to determine the parameters $\epsilon$, $\tau$, and $\beta$, the efficency, deadtime, and dieaway time of our detection system. The interval size used in neutron fluctuation measurements is optimized to produce a minimum fractional statistical error for a fixed counting time in Q, the measurement value that is proportional to the time correlated neutron count rate from the sample.

## 2. Theory

We consider a system in which neutrons are produced by ($\alpha$,n) reactions, spontaneous fission and neutron induced fission. We use the following forms for $P(t)$ and $P_f(t)$; the probability that a neutron born at time 0 produces a pulse in dt about t and the probability that a neutron born at time 0 produces a fission in dt about t, respectively

$$P(t)dt = \epsilon \beta e^{-\beta t} dt$$

$$P_f(t)dt = \frac{k_p \beta}{\bar{\nu}_1} e^{-\alpha t} dt$$

where: $\varepsilon$ = detector efficiency (assumed independent of neutron origin)
$\beta$ = inverse neutron lifetime
$\alpha$ = Rossi-alpha = $(1-k_p)\beta$
$k_p$ = prompt neutron multiplication factor
$\bar{\nu}_1$ = mean number of neutrons per induced fission

Let $P_n(t_1, t_2, \ldots, t_n)dt_1, dt_2, \ldots, dt_n$ be the probability of detecting n pulses occurring in $dt_1$ about $t_1$, $dt_2$ about $t_2$, ... and $dt_n$ about $t_n$, then

$$P_1(t_1) = \varepsilon S$$

$$P_2(t_1,t_2) = \varepsilon^2 S^2 + \varepsilon^2\, \overline{\nu(\nu-1)}\, F\, \frac{\beta e^{-\alpha(t_2-t_1)}}{2(1-k_p)}$$

$$P_3(t_1,t_2,t_3) = \varepsilon^3 S^3 + \frac{\varepsilon^3 S\overline{\nu(\nu-1)}F\alpha}{2(1-k_p)^2}\left[e^{-\alpha(t_3-t_1)} + e^{-\alpha(t_3-t_2)} + e^{-\alpha(t_2-t_1)}\right]$$

$$+ \frac{\varepsilon^3\overline{\nu(\nu-1)(\nu-2)}F\alpha}{3(1-k_p)^3}\, e^{-\alpha(t_2-t_3-2t_1)}$$

$$+ \frac{\varepsilon^3\overline{\nu(\nu-1)}Fk_1\overline{\nu_1(\nu_1-1)}\alpha^2}{2\bar{\nu}_1(1-k_1)^4}\, e^{-\alpha(t_3-t_1)}$$

where: $S$ = total source strength $= (R+\bar{\nu}_o F_o+\bar{\nu}_1 F_1)$
$R$ = uncorrelated $(\alpha,n)$ source strength
$\bar{\nu}_o F_o$ = spontaneous fission source strength
$\bar{\nu}_1 F_1$ = neutron induced fission source strength

and we have used the shorthand

$$\overline{\nu(\nu-1)}F = \overline{\nu_o(\nu_o-1)}F_o + \overline{\nu_1(\nu_1-1)}F_1$$

$$\overline{\nu(\nu-1)(\nu-2)}F = \overline{\nu_o(\nu_o-1)(\nu_o-2)}F_o + \overline{\nu_1(\nu_1-1)(\nu_1-2)}F_1$$

If the detection system deadtime after a pulse is $\tau$, then the expected number of counts in a time channel of width $T_o$ is

$$\bar{C} = \int_o^{T_o} dt_1 P_1(t_1)\, e^{-\int_{t_1-\tau}^{t_1} W_1(t_1,t_2)dt_2}$$

where $W_1(t_1,t_2)dt_2$ is the conditional probability that given a pulse at $t_1$, there is also a pulse in $dt_2$ about $t_2$, and is given by the relation

$$P_2(t_1,t_2) = P_1(t_1)W(t_1,t_2)$$

For small deadtime, we make the approximation

$$e^{-\int_{t_1-\tau}^{t_1} W(t_1,t_2)dt_2} \doteq 1 - \int_{t_1-\tau}^{t_1} W_1(t_1,t_2)dt_2$$

and obtain:

$$\bar{C} = \int_o^{T_o-\tau} dt_1\left[P_1(t_1) - \int_{t_1-\tau}^{t_1} dt_2 P_2(t_1,t_2)dt_2\right]$$

Similarly:

$$\frac{\overline{C(C-1)}}{2} = \int_o^{T_o-\tau} dt_1 \int_{t_1+\tau}^{T_o} dt_2 P_2(t_1,t_2)\; e^{-\left[\int_{t_1-\tau}^{t_1} dt_3 W_2 + \int_{t_2-\tau}^{t_2} dt_3 W_2\right]}$$

where $W_2(t_1,t_2,t_3)dt_3$ is the conditional probability that, given a pulse pair at $t_1, t_2$, there is also a pulse in $dt_3$ about $t_3$ and is given by the relation:

$$P_3(t_1,t_2,t_3) = P_2(t_1,t_2)W_2(t_1,t_2,t_3)$$

Then using an approximation similar to that used above for the case of small deadtime, we obtain:

$$\frac{\overline{C(C-1)}}{2} = \int_o^{T_o-\tau} dt_1 \int_{t_1+\tau}^{T_o} dt_2 \left[P_2(t_1,t_2) - \int_{t_1-\tau}^{t_1} P_3(t_1,t_2,t_3)dt_3 - \int_{t_2-\tau}^{t_2} P_3(t_1,t_2,t_3)dt_3\right]$$

Finally, we obtain:

$$\bar{C} = \varepsilon S T_o\left[1 - \varepsilon\tau\left\{S + \frac{\overline{\nu(\nu-1)}F\beta}{2S(1-k_p)}\right\}\right]$$

and

$$\overline{C(C-1)} = a_1 T^2 + 2a_2 e^{-\alpha\tau}\frac{T}{\alpha}\left[1 - \frac{1-e^{-\alpha T}}{\alpha T}\right]$$

$$+ a_3 e^{-2\alpha T}\frac{T}{\alpha}\left[1 - \frac{1-e^{-2\alpha T}}{2\alpha T}\right]$$

where:

$$T = T_o - \tau$$

$$a_1 = \epsilon^2 S^2 - 2\epsilon^3 S^3 \tau - \frac{\epsilon^3 S \overline{\nu(\nu-1)} F \alpha \tau}{(1-k_p)^2}$$

$$a_2 = \frac{\epsilon^3 \overline{\nu(\nu-1)} F \alpha}{2(1-k_p)^2} - \frac{2\epsilon^3 \tau S \overline{\nu(\nu-1)} F \alpha}{(1-k_p)^2}$$

$$- \frac{\epsilon^3 \tau \overline{\nu(\nu-1)(\nu-2)} F \alpha^2}{3(1-k_p)^3} - \frac{\epsilon^3 \overline{\nu(\nu-1)} F k_p \overline{\nu_1(\nu_1-1)} \alpha^2}{\bar{\nu}_1 (1-k_p)^4}$$

$$a_3 = - \frac{\epsilon^3 \tau \overline{\nu(\nu-1)(\nu-2)} F \alpha^2}{3(1-k_p)^3}$$

Using the approximations:

$$\left(1 - \frac{T^2}{T_o^2}\right) \doteq \frac{2\tau}{T_o}$$

and

$$a_1 \doteq \frac{\bar{C}^2}{T_o^2}$$

we obtain:

$$\overline{C(C-1)} - \bar{C}^2 + \frac{2\tau}{T_o} \bar{C}^2 = \frac{\epsilon^2 \overline{\nu(\nu-1)} F T g(\alpha T) e^{-\alpha\tau}}{(1-k_p)^2} \left[1-4\tau h\right]$$

where:

$$g(x) = 1 - \frac{1-e^{-x}}{x}$$

$$h = \epsilon S + \frac{\epsilon F}{6} \frac{\overline{\nu(\nu-1)(\nu-2)}}{\overline{\nu(\nu-1)}} \left[1 + e^{-\alpha T} \frac{g(2\alpha T)}{2g(\alpha T)}\right]$$

$$+ \frac{\epsilon F k_p \overline{\nu_1(\nu_1-1)}}{2\bar{\nu}_1 (1-k_p)} \approx \epsilon S + \frac{\epsilon F k_p \overline{\nu_1(\nu_1-1)}}{2\bar{\nu}_1 (1-k_p)}.$$

The approximation results from neglecting the term due to counting of triples from a fission event.

Letting $\overline{C(C-1)} - \bar{C}^2 = Q_m$, one has

$$Q_m + \frac{2\tau}{T_o} \bar{C}^2 = \frac{\epsilon^2 \left\{ \overline{\nu_o(\nu_o-1)}\ F_o + \overline{\nu_1(\nu_1-1)}\ F_1 \right\}}{(1-k_p)^2}$$

$$\times\ T g(\alpha T) e^{-\alpha\tau} \left[1-4\tau h\right]$$

An examination of this expression reveals that the multiplication, $M = 1/(1-k_p)$, and the spontaneous and induced fission components $F_o$ and $F_1$ are inseparable without knowledge of two of the three unknowns $M$, $F_o$, and $F_1$.

However, for the special case of pure metal samples where there are negligible $(\alpha,n)$ sources, we can make the substitutions

$$\bar{C}_o = \bar{C}/M \text{ and } \bar{C}_1 = \bar{C}(M-1)/M$$

and obtain

$$Q_m + \frac{2\tau}{T_o} \bar{C}^2 = \epsilon M \bar{C} \left\{ D_o + (M-1) D_1 \right\}$$

$$\times \frac{T}{T_o}\ g(\alpha T) e^{-\alpha\tau} \left[1-4\tau H\right]$$

where $D_i = \overline{\nu_i(\nu_i-1)} / \bar{\nu}_i$ and

$$H \simeq \frac{3\bar{C}}{4T_o} + \frac{\epsilon\beta}{8} \left\{ 3(M-1)D_1 - D_o \right\}$$

All of the variables and parameters in this expression are known or measured except for the multiplication $M$ which is thence determined by observation of $Q_m$. The count rate, $\bar{C}_o$, which is proportional to the effective $^{240}$Pu mass, is thence determined from $\bar{C}_o = \bar{C}/M$.

For another special case of a nonmultiplying sample containing $(\alpha,n)$ sources, we set $k_p = 0$, and obtain

$$Q_m + \frac{2\tau \bar{C}^2}{T_o} = \epsilon \bar{C}_o D_o \frac{T}{T_o}\ g(\beta T) e^{-\beta\tau} \left[1-4\tau H'\right]$$

where

$$H' = \frac{3\bar{C}}{4T_o} - \frac{\epsilon\beta}{8} \frac{D_o \bar{C}_o}{\bar{C}}$$

This expression can be solved directly for $\bar{C}_o$.

## 3. Experimental Method

The system that is used for our investigations of the reduced variance method consists of a typical large (60cm x 60cm x 70cm) polythene-moderated well type neutron detector with sixteen 2.5-cm diameter, 60-cm long $^3$He proportional counters, associated logic pulse generating electronics, and a microNOVA computer interfaced to the electronics via scaler/timer interface boards. Total measurement times and individual time bin widths can be user selected via a keyboard or may be placed under software control for parametric measurements. The reduced variance algorithm calls for the accumulation of the complete neutron pulse distribution and the calculation of the first through fourth moments of the distribution about the origin and the combinations of these moments required for the error analysis of $Q_m$.

## 4. Results

The data in Table I were obtained assuming that $\bar{C}_0$, the average number of counts per interval due to fission, does not charge when additional random ($\alpha$,n) source neutrons are added to the system. These data were fitted by weighted least squrres with the expression for $Q_m$ above using the value $\bar{\nu}_0 = 3.74$ and $\bar{\nu}_n = 15.54$ for $^{252}Cf$ to obtain:

$$\epsilon = 0.1594 \pm 0.0019$$
$$\tau = 1.332 \pm 0.0095\ \mu s$$
$$\beta = 0.01488 \pm 0.00047\ \mu s^{-1}$$

A very interestir result of this analysis routine is that the efficiency of the neutron detector has been determined without the use of calibration standards.

For any measurement of Q, the standard deviation of Q, $\sigma_Q$, can be readily determined using the moments:

$$\sigma^2_{\bar{C}} = \frac{\overline{C^2} - \bar{C}^2}{N-1}$$

$$\sigma^2_{\overline{C^2}} = \frac{\overline{C^4} - (\overline{C^2})^2}{N-1}$$

$$\sigma_{\bar{C}\,\overline{C^2}} = \frac{\overline{C^3} - \overline{C^2}\bar{C}}{N-1}$$

where N is the number of count intervals in the measurement and the variances and covariances of $\epsilon$, $\tau$, and $\beta$ are available from the least squares fit above:

$$\sigma_{\tau\beta} = 1.363 \times 10^{-6}$$
$$\sigma_{\tau\epsilon} = 5.599 \times 10^{-6}$$
$$\sigma_{\epsilon\beta} = -0.6013 \times 10^{-6}$$

It is assumed that covariances between $\bar{C}$, $\overline{C^2}$, and $\tau$, $\epsilon$, $\beta$ are zero because these sets of values are obtained from different measurements.

Measurements of Q were made with a $^{252}Cf$ source and a constant run time (2000 s) with interval size $T_0$ varied over a wide range. A plot of the ratio $\sigma_Q/Q$ is given in Fig. 1. This shows an optimum interval size of approximately 150$\mu$s, on the order of twice the dieaway time.

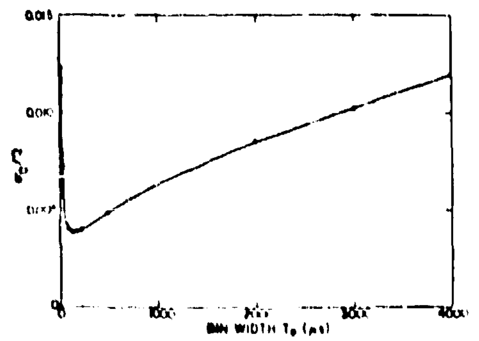

Fig. 1

Again, using the detector system for which the parameters $\tau$, $\epsilon$, and $\beta$ have been determined, and the expressions for $Q_m$, the effective $^{240}Pu$ mass and multiplication can be determined for pure metal samples, and the effective $^{240}Pu$ mass can be determined for any nonmultiplying sample.

## 5. Conclusions

The complete formalism for the determination, using the reduced variance or neutron fluctuation method, of the fission correlated pulses in a pulse train from neutron detectors has been developed. The formalism explicitly includes the effects of detector efficiency, deadtime and dieaway time, and contributions from ($\alpha$,n) and induced fission events. An error estimator has also been provided. The formalism has been used to completely characterize a neutron well counter used for plutonium sample assays. We have shown how the effective $^{240}Pu$ mass and multiplication can be determined for metal plutonium samples without the parametric method invoked in our earlier paper, and how the effective $^{240}Pu$ mass can be determined for any nonmultiplying sample containing ($\alpha$,n) sources. We have not yet used the formalism for the determination of $^{240}Pu$ mass in multiplying samples containing ($\alpha$,n) sources.

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TABLE I

MEASUREMENT DATA USED IN DETERMINING THE PARAMETERS IN THE EXPRESSION FOR $Q_m$

| Source | $T_o$, μs | $\bar{C}_o$ | $\bar{C}$ | $Q_m$ | $Q_E$ |
|---|---|---|---|---|---|
| Cf-1 | 100 | 0.02520 | 0.02520 | 0.00570 | 0.00008 |
| | 200 | 0.0500 | 0.0500 | 0.0161 | 0.0001 |
| | 500 | 0.1248 | 0.1248 | 0.0506 | 0.0003 |
| | 1000 | 0.2488 | 0.2488 | 0.1079 | 0.0008 |
| | 2000 | 0.496 | 0.496 | 0.224 | 0.002 |
| | 4000 | 0.994 | 0.994 | 0.456 | 0.005 |
| Cf-1 + (α,n) | 100 | 0.02520 | 0.0600 | 0.00578 | 0.00005 |
| | 200 | 0.0500 | 0.1198 | 0.0160 | 0.0001 |
| | 500 | 0.1248 | 0.2966 | 0.0508 | 0.0005 |
| | 1000 | 0.2488 | 0.594 | 0.110 | 0.001 |
| | 2000 | 0.496 | 1.188 | 0.23 | 0.01 |
| | 4000 | 0.994 | 2.374 | 0.449 | 0.008 |
| Cf-2 | 100 | 0.2017 | 0.2017 | 0.0454 | 0.0009 |
| | 200 | 0.3992 | 0.3992 | 0.1281 | 0.0004 |
| | 500 | 0.9958 | 0.9958 | 0.400 | 0.002 |
| | 1000 | 1.988 | 1.988 | 0.859 | 0.004 |
| | 2000 | 3.975 | 3.975 | 1.77 | 0.01 |
| | 4000 | 7.945 | 7.945 | 3.59 | 0.03 |
| Cf-2 + (α,n) | 100 | 0.2017 | 2.9117 | -0.197 | 0.001 |
| | 200 | 0.3992 | 5.78 | -0.361 | 0.003 |
| | 500 | 0.9958 | 14.404 | -0.83 | 0.02 |
| | 1000 | 1.988 | 28.767 | -1.62 | 0.04 |
| | 2000 | 3.975 | 57.49 | -3.2 | 0.1 |
| | 4000 | 7.945 | 114.89 | -6.0 | 0.3 |
| Cf-3 | 100 | 0.4364 | 0.4364 | 0.0945 | 0.0003 |
| | 200 | 0.8675 | 0.8675 | 0.2693 | 0.0008 |
| | 500 | 2.160 | 2.160 | 0.856 | 0.003 |
| | 1000 | 4.307 | 4.307 | 1.839 | 0.009 |
| | 2000 | 8.624 | 8.624 | 3.81 | 0.03 |
| | 4000 | 17.23 | 17.23 | 7.86 | 0.07 |
| Cf-3 + (α,n) | 100 | 0.4364 | 3.1178 | -0.190 | 0.001 |
| | 200 | 0.8675 | 6.19 | -0.312 | 0.004 |
| | 500 | 2.160 | 15.421 | -0.62 | 0.01 |
| | 1000 | 4.307 | 30.80 | -1.19 | 0.04 |
| | 2000 | 8.624 | 61.6 | -2.5 | 0.1 |
| | 4000 | 17.23 | 123.06 | -4.5 | 0.3 |
| Cf-4 | 100 | 4.3171 | 4.3171 | 0.294 | 0.002 |
| | 200 | 8.577 | 8.577 | 1.252 | 0.006 |
| | 500 | 21.348 | 21.348 | 4.66 | 0.03 |
| | 1000 | 42.655 | 42.655 | 10.26 | 0.07 |
| | 2000 | 85.21 | 85.21 | 21.5 | 0.2 |
| | 4000 | 170.26 | 170.26 | 44.0 | 0.6 |
| Cf-4 + (α,n) | 100 | 4.3171 | 6.6776 | -0.556 | 0.003 |
| | 200 | 8.577 | 13.264 | -0.558 | 0.008 |
| | 500 | 21.348 | 33.015 | -0.14 | 0.03 |
| | 1000 | 42.655 | 65.96 | 0.64 | 0.09 |
| | 2000 | 85.21 | 131.81 | 2.3 | 0.3 |
| | 4000 | 170.26 | 263.41 | 5.9 | 0.8 |
| Cf-5 | 100 | 6.902 | 6.902 | -0.185 | 0.003 |
| | 200 | 13.711 | 13.711 | 0.565 | 0.009 |
| | 500 | 34.132 | 34.132 | 3.51 | 0.04 |
| | 1000 | 68.17 | 68.17 | 8.5 | 0.1 |
| | 2000 | 136.27 | 136.27 | 18.5 | 0.3 |
| | 4000 | 272.29 | 272.29 | 38.2 | 0.9 |
| Cf-5 + (α,n) | 100 | 6.902 | 9.0958 | -1.342 | 0.003 |
| | 200 | 13.711 | 18.071 | -1.89 | 0.01 |
| | 500 | 34.132 | 44.991 | -2.86 | 0.04 |
| | 1000 | 68.17 | 89.86 | -4.4 | 0.1 |
| | 2000 | 136.27 | 179.57 | -7.3 | 0.4 |
| | 4000 | 272.29 | 358.91 | -13.9 | 0.9 |
| (α,n) | 100 | 0.0 | 2.7020 | -0.206 | 0.001 |
| | 200 | 0.0 | 5.368 | -0.408 | 0.003 |
| | 500 | 0.0 | 13.366 | -1.02 | 0.01 |
| | 1000 | 0.0 | 26.695 | -2.05 | 0.03 |
| | 2000 | 0.0 | 53.34 | -3.9 | 0.1 |
| | 4000 | 0.0 | 106.62 | -8.0 | 0.3 |